

## Article

# Experimental and Simulation Research on the Process of Nitrogen Migration and Transformation in the Fluctuation Zone of Groundwater Level

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**Abstract:** The fluctuation of groundwater causes a change in the groundwater environment and then affects the migration and transformation of pollutants. To study the influence of water level fluctuations on nitrogen migration and transformation, physical experiments on the nitrogen migration and transformation process in the groundwater level fluctuation zone were carried out. A numerical model of nitrogen migration in the Vadose zone and the saturated zone was constructed by using the software HydrUS-1D. The correlation coefficient and the root mean square error of the model show that the model fits well. The numerical model is used to predict nitrogen migration and transformation in different water level fluctuation scenarios. The results show that, compared with the fluctuating physical experiment scenario, when the fluctuation range of the water level increases by 5 cm, the fluctuation range of the nitrogen concentration in the coarse sand, medium sand and fine sand media increases by 37.52%, 31.40% and 21.14%, respectively. Additionally, when the fluctuation range of the water level decreases by 5 cm, the fluctuation range of the nitrogen concentration in the coarse sand, medium sand and fine sand media decreases by 36.74%, 14.70% and 9.39%, respectively. The fluctuation of nitrogen concentration varies most significantly with the amplitude of water level fluctuations in coarse sand; the change in water level has the most significant impact on the flux of nitrate nitrogen and has little effect on the change in nitrite nitrogen and ammonium nitrogen, and the difference in fine sand is the most obvious, followed by medium sand, and the difference in coarse sand is not great.

**Keywords:** groundwater level fluctuation zone; nitrogen; migration and transformation; HYDRUS-1D model



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## 1. Introduction

Due to industry [1,2], agriculture [3,4], life [5], aquaculture [6], atmospheric deposition [7] and other factors, the groundwater in many countries and regions around the world are affected by nitrogen pollution, including the United States [8], China [9], the United Kingdom [10], South Korea [11] and other countries with severe pollution. In 2005, 34.1% of 1139 groundwater samples in northern China failed to meet World Health Organization (WHO) criteria. [12]. Currently, China has become one of the countries with the highest nitrogen fertilizer used in the world [13]. Nitrate from nitrogen may rapidly leach into groundwater, affecting the ecology and human health [14]. Wild et al. concluded that even if nitrate input is significantly reduced in the future, it will take decades to significantly reduce nitrate concentrations in porous aquifers through denitrification [15]. In 2000, the Water Framework Directive to bring water bodies to good chemical and ecological status by 2027 was issued [16]. Therefore, how to scientifically understand the nitrogen migration and transformation patterns under fluctuating groundwater level conditions has become one of the hot issues in the field of environmental research.

During groundwater level fluctuations, soluble substances in the aquifer medium are gradually dissolved into the groundwater, thus changing the chemical composition of the groundwater [17]. Guo Huaming et al. [18] studied the influencing factors of arsenic enrichment in groundwater, and the results showed that a high pH was unfavorable to the adsorption of arsenic in the form of anions by the aquifer medium. In their study, Sorensen J et al. [19] found that chemical and biological contaminants near the surface were transported to groundwater with minimal attenuation. Water level fluctuations also have a significant effect on nitrogen. Hefting M et al. [20] selected 13 riparian sites to analyze nitrogen cycling processes and confirmed a direct positive correlation between denitrification and elevated water table levels. Heather L. Welch et al. [21] analyzed and found that nitrate-N is weakened by denitrification during downward transport using redox sensitivity metrics at the water table without a location in the vertical direction. Jurado A et al. [22] concluded that the accumulation of  $N_2O$  in groundwater is also mainly due to denitrification and, to a lesser extent, nitrification. Yang L.P. et al. [23] conducted indoor soil column experiments to study the effect of pH on nitrogen transport and transformation processes and showed that pH 6.5 was the most efficient for the removal of “tri-nitrogen” in the optimal pH range for adsorption nitrification and denitrification. The authors in [24] analyzed the effect of water level fluctuation on nitrogen transformation by simulating aerobic, anoxic and anaerobic zones, and the results showed that denitrification played the greatest role in the anaerobic zone.

Groundwater level fluctuations significantly affect the migration and transformation of groundwater pollutants [25–29]. Scholars at home and abroad have conducted studies on groundwater level fluctuations. Most of their research focuses on the migration and transformation processes of soil salinity, iron, manganese, arsenic, iodine, benzene and other characteristic components when subjected to water level fluctuations, etc. [18,30,31]. Davis et al. [32] studied the changes of organic matter and oxygen at the fluctuation of water level, and the results showed that BTEX and oxygen concentration showed a relationship between this and that near the water level, and the main reason for oxygen reduction was due to microbial degradation. Kamon et al. [33] conducted an experimental and numerical study of the migration of LNAPL with water level fluctuations and showed that the entry and displacement pressures were greater for the air-water system than for the LNAPL-water system. Xiaoxi Xie et al. [34] analyzed the hysteresis relationship between saturation and the capillary pressure in a medium under the water table fluctuation conditions during alternating drying and wetting processes, and the results demonstrated that when the initial water saturation of the drying process is similar, the greater the initial water saturation of the wetting process, the degree of hysteresis gradually decreases. Xiang Li et al. [35] analyzed and verified that groundwater level fluctuations affected the physicochemical properties of soil-water bodies and further affected the movement of nitrate in soil solids.

Research on the mechanisms of solute migration and transformation in the fluctuation zone of groundwater level, mainly through technical methods, such as field investigation, indoor experiments, theoretical analysis and numerical simulation, was conducted to predict the temporal and spatial distribution characteristics of solute pollution. Chen et al. [36] found through field experiments, that the depth of shallow groundwater has more influence on the nitrogen concentration in shallow groundwater than other factors. Zhang Dan et al. [27] conducted on-site monitoring of plots at different altitudes for a year. They found that the fluctuation of shallow groundwater levels significantly affected the soil profile and the nitrogen concentration of shallow groundwater. Farnsworth et al. [37] established an indoor soil column experiment, using a set of 1.3 m quartz sand columns inoculated with microorganisms, and changing the water level in the sand column every 30–50 h to simulate the periodic production wells of production wells and groundwater caused by the cessation of mining and the influence of the level fluctuation on the oxidation of manganese—the content of manganese increases as the water level drops, and decreases as the water levels rise. Yang Yang [38] studied the influence of the groundwater level rise and fall on the migration of cadmium, and argued that the rise and fall in the water level

mainly affect the transport of cadmium ions through convection. Zhang Xuejing et al. [39] used inverse distance weighting (IDW) interpolation and the water chemistry Piper graphic method to analyze the response relationship between the groundwater chemistry characteristics and the depth of the water level in the Ejina Oasis after the ecological water transport (2001–2017). Cao Wengeng et al. [40] used the potential distribution-multi-point complexation model (CD-MUSIC) to predict the evolution of groundwater chemical composition and hydrochemical types in the Baoding Plain of the South-to-North Water Transfer Project under the condition of groundwater level rebound. Arash Taftah and Ali Reza Sepaskhah [41] successfully simulated the leaching of water and nitrate from two crops in the field with high accuracy using HYDRUS-1D. Mo Xiaoyu et al. [42] used HYDRUS-1D to simulate the changes in nitrogen leaching under different rainfall intensities, analyzed the influencing factors, and found that high intensity would reduce the nitrogen utilization rate.

In summary, the impact of water level fluctuations on the migration and transformation of pollutants has attracted the attention of scholars. Still, the study of nitrogen migration and transformation under different media water level fluctuations is not systematic enough. In this paper, the indoor soil column is used to simulate water level fluctuations to study the temporal and spatial distribution of nitrate nitrogen, nitrite nitrogen and ammonium nitrogen in three typical soil media subjected to water level fluctuations. According to physical experimental conditions, a numerical model of nitrogen migration and transformation in the fluctuation zone of the groundwater level was established to predict the spatial distribution and temporal change of nitrogen pollutants in the fluctuation zone of the groundwater level under different scenarios. This paper provides a scientific basis for the treatment, restoration and protection of groundwater nitrogen pollution.

## 2. Materials and Methods

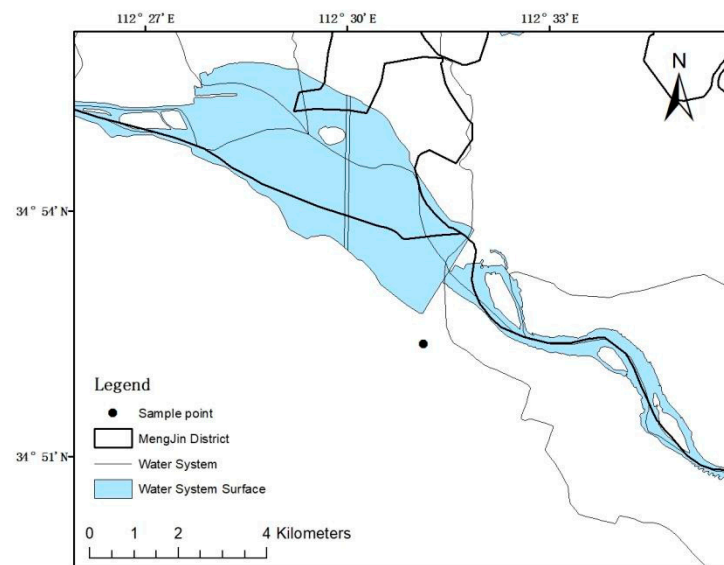
### 2.1. Experimental Design

#### 2.1.1. Experimental Materials

The experimental soil samples were collected on the floodplain of the Yellow River in Mengjin District, Luoyang City, Henan Province (Figure 1). The groundwater level in this area fluctuates frequently. Sample collection and processing were carried out by the requirements of the “Technical Specifications for Soil Environmental Monitoring” (HJ/T 166-2004). Sampling was performed using the quarter method, followed by drying, crushing and sieving, delineating three media, namely coarse sand, medium sand and fine sand. The pH value of the sand was between 8.5 and 9.3, and the organic matter content was between 0.241 and 1.070 g·kg<sup>-1</sup>. The basic physical and chemical properties of the soil are shown in Table 1. The water used in the laboratory is ultrapure water made by the German Millipore ultrapure water machine.

#### 2.1.2. Experimental Device

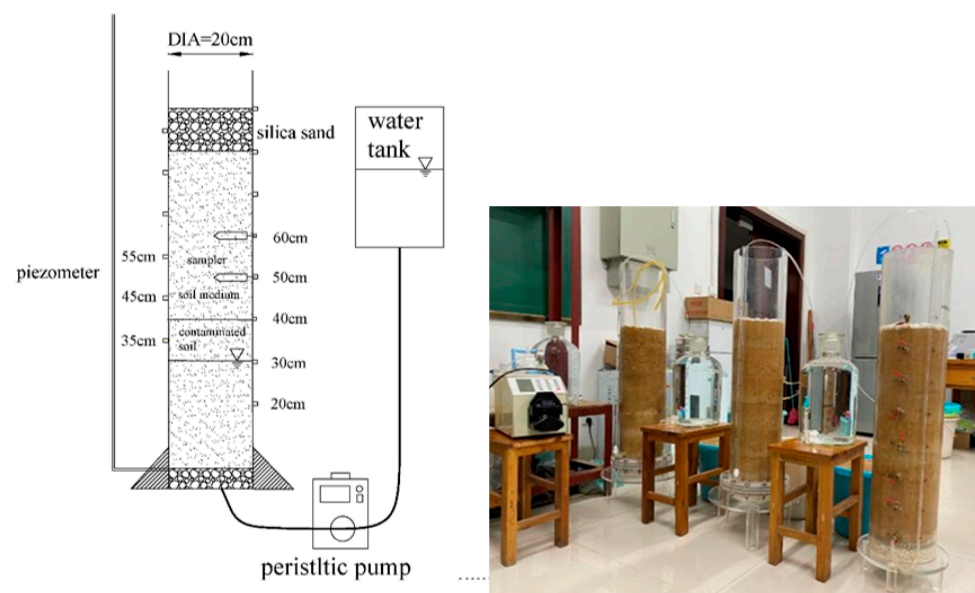
The experiment designed three plexiglass columns with the following exact specifications: the inner diameter was 20 cm, the height was 110 cm, the top was open, and the opening of the glass column was covered with plastic film; the bottom was connected to a pressure-measuring tube and a Markov flask through a three-way valve. Between the Markov flask and the cylinder, a peristaltic pump was equipped to control the water level to simulate the process of groundwater fluctuation (rising–falling); the organic glass column was equipped with four sampling ports from top to bottom, at 15, 25, 35, and 45 cm away from the bottom of the column; adjacent points were separated by 10 cm. The sampling points were equipped with a Rhizon solution sampler (inner diameter: 1 mm); we covered the top and bottom of each column with 5 cm of quartz sand (diameter 2–3 mm), to ensure that the water level rose and fell uniformly. The experimental device is shown in Figure 2.



**Figure 1.** Location of sampling points.

**Table 1.** Soil particle composition and its parameters.

Serial Number	Sample	Particle Size (mm)	pH	Organic Matter Content ( $\text{g}\cdot\text{kg}^{-1}$ )
1	Coarse sand	0.5–1.0	8.5	0.241
2	Medium sand	0.25–0.5	8.8	0.587
3	Fine sand	0.125–0.25	9.3	1.070



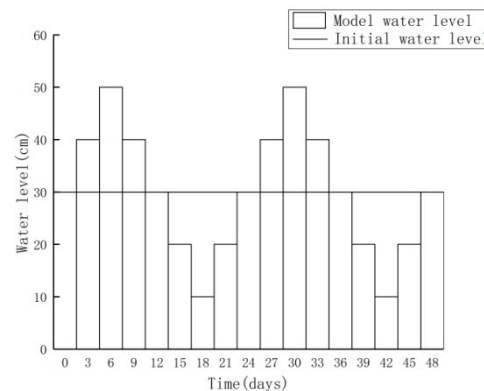
**Figure 2.** Schematic diagram of the experimental device.

### 2.1.3. Experimental Method

First, the experiments were conducted using soil from different media to fill the columns, filled with distilled water and stabilized at a 30 cm scale at the initial water level. Then, potassium nitrate and ammonium chloride solutions were injected from the top of the column. Three days later, we took out the water sample, and adjusted the water level using a peristaltic pump, raising its height by 10 cm each time until the water level reached 50 cm; after that, we decreased the water level by 10 cm each time, until the water level



dropped to 10 cm. Finally, we raised the water level again, by 10 cm each time, until it reached 30 cm. The water levels during the experimental period were 30, 40, 50, 40, 30, 20, 10, 20, and 30 cm, respectively. Each water level was maintained for 3 d, and the change in the water level over time is shown in Figure 3, with the experiment using 24 d as a complete change period. The investigation was carried out for two cycles.



**Figure 3.** Time variation of water level.

Before each water level change, water samples were collected at four sampling ports using a diaphragm vacuum pump and a soil solution sampler to measure the concentrations of three forms of ammonium nitrogen, nitrate nitrogen and nitrite nitrogen.

## 2.2. Numerical Model

HYDRUS-1D is widely used in saturated-unsaturated zones of water, heat and solute transport to study the process of nitrogen migration and the transformation in the fluctuation zone at the groundwater level. In this paper, based on indoor physical experiments, a numerical model of nitrogen migration and change in the Vadose zone–saturated zone coupled with water transport and solute transport, was constructed using HYDRUS-1D.

### 2.2.1. Mathematical Model

- **Water Movement Model.**

The mathematical model of water movement in the water level fluctuation zone can be expressed as the Richards equation, as follows:

$$C(h) \frac{\partial \theta}{\partial t} = \frac{\partial}{\partial z} \left[ K(h) \left[ \frac{\partial h}{\partial t} - \cos(\alpha) \right] \right] - S(z, t) \quad (1)$$

where  $C(h)$  is the water capacity ( $\text{cm}^{-1}$ );  $K(h)$  is the hydraulic conductivity ( $\text{cm}/\text{d}$ );  $h$  is the negative pressure ( $\text{cm}$ );  $z$  represents the position coordinates in the parallel water flow direction ( $\text{cm}$ );  $t$  is the time ( $\text{d}$ );  $\alpha$  is the angle between the water flow direction and the vertical ( $^\circ$ );  $\theta$  is the volumetric water content ( $\text{cm}^3/\text{cm}^3$ ); and  $S(z, t)$  is the water absorption strength of plant roots ( $\text{cm}^3/\text{cm}^3 \cdot \text{d}^{-1}$ ).

- **Solute transport model in the Vadose zone.**

The solute transport model only considers the behavioral characteristics of convection, diffusion, adsorption, degradation, etc., and uses the traditional convection–diffusion equation to describe the transport process. The equation is expressed as:

$$\frac{\partial}{\partial t}(\theta C) = \frac{\partial}{\partial z} \left( \theta D_L \frac{\partial C}{\partial z} - vC \right) - \rho_b \frac{\partial (\rho_s K_L C)}{\partial t} - C_0 \exp(-kt) \quad (2)$$

where  $C$  is the solute concentration in the liquid phase ( $\text{mg}/\text{L}$ );  $D_L$  is the longitudinal dispersion coefficient ( $\text{cm}/\text{d}$ );  $v$  is the Darcy flow velocity ( $\text{cm}/\text{d}$ );  $\rho_b$  is the soil bulk density ( $\text{mg}/\text{cm}^3$ );  $\rho_s$  is the soil bulk density ( $\text{mg}/\text{cm}^3$ );  $K_L$  is the adsorption distribution

coefficient ( $\text{cm}^3/\text{g}$ );  $C_0 \exp(-kt)$  is the source-sink term ( $\text{cm}^3/\text{cm}^3 \cdot \text{d}^{-1}$ ); the others are the same as above.

- Nitrogen migration and transformation model.

The transport model of nitrogen in the soil varies according to the different forms of nitrogen. The transport and transformation process of  $\text{NH}_4^+$  is mainly subjected to adsorption and nitrification. The equation is as follows:

$$\begin{cases} \frac{\partial(\theta c_1)}{\partial t} = \frac{\partial}{\partial z} \left( \theta D_L \frac{\partial c_1}{\partial z} \right) - \frac{\partial}{\partial z} (V c_1) - \rho_b \frac{\partial(\rho_s k_e c_1)}{\partial t} - k_1 \theta c_1 \\ c_1(Z, 0) = c_{10}(Z) & 0 \leq Z \leq L, t = 0 \\ - \left( \theta D_L \frac{\partial c_1}{\partial z} - q c_1 \right) = \varepsilon(t) c_0 & Z = 0, t = 0 \\ c_1(L, t) = c_{1L} & Z = L, t > 0 \end{cases} \quad (3)$$

Meanwhile  $\text{NO}_2^-$  and  $\text{NO}_3^-$  are mainly affected by nitrification and denitrification, and the equation is as follows:

$$\begin{cases} \frac{\partial(\theta c_i)}{\partial t} = \frac{\partial}{\partial z} \left( \theta D_L \frac{\partial c_i}{\partial z} \right) - \frac{\partial}{\partial z} (v_i c_i) - k_i \theta c_i \\ c_i(Z, 0) = c_{i0}(Z) & 0 \leq Z \leq L, t = 0 \\ c_i(0, t) = c_{i0}(t) & Z = 0, t > 0 \\ c_i(L, t) = c_{iL} & Z = L, t > 0 \end{cases} \quad (4)$$

where  $\theta$  is the soil volume water content ( $\text{cm}^3/\text{cm}^3$ );  $c_1$  is the soil solution  $\text{NH}_4^+$  concentration ( $\text{mg}/\text{L}$ );  $D_L$  is the longitudinal diffusion coefficient ( $\text{cm}/\text{d}$ );  $k_e$  is the adsorption distribution coefficient ( $\text{cm}^3/\text{g}$ ) for  $\text{NH}_4^+$  in soil;  $k_1, k_2$  are the denitrification rate constants ( $\text{d}^{-1}$ ) for  $\text{NH}_4^+$  and  $\text{NO}_2^-$ , respectively;  $c_{10}(t)$  is the soil  $\text{NH}_4^+$  initial concentration ( $\text{mg}/\text{L}$ );  $c_0$  is the inlet solution  $\text{NH}_4^+$  concentration ( $\text{mg}/\text{L}$ );  $c_{1L}$  is the diving  $\text{NH}_4^+$  concentration ( $\text{mg}/\text{L}$ );  $c_2, c_3$  are the  $\text{NO}_2^-$  and  $\text{NO}_3^-$  concentration ( $\text{mg}/\text{L}$ ), respectively;  $k_3$  is the denitrification rate constant ( $\text{d}^{-1}$ );  $c_{20}(z), c_{30}(z)$  are the soil  $\text{NO}_2^-$  and  $\text{NO}_3^-$  initial concentration ( $\text{mg}/\text{L}$ ), respectively; and  $c_{2L}, c_{3L}$  are the diving  $\text{NO}_2^-$  and  $\text{NO}_3^-$  concentration ( $\text{mg}/\text{L}$ ), respectively. No nitrogen input through the water.

## 2.2.2. Initial Conditions and Boundary Conditions

- Initial conditions.

At the initial moment, the water level was set to 30 cm, and the initial concentration of pollutants is shown in Table 2.

**Table 2.** Initial concentration of pollutants.

Intake	Coarse Sand			Medium Sand			Fine Sand		
	Nitrite Nitrogen	Nitrate	Ammonium Nitrogen	Nitrite Nitrogen	Nitrate	Ammonium Nitrogen	Nitrite Nitrogen	Nitrate	Ammonium Nitrogen
15 cm	0.19	74.85	6.17	0.37	35.81	1.97	0.70	0.55	0.08
25 cm	0.15	77.14	6.97	0.27	36.47	3.07	0.84	2.20	0.09
35 cm	0.37	47.56	7.03	0.36	78.39	11.87	0.44	12.61	5.14
45 cm	0.33	45.25	8.23	0.25	84.41	12.61	0.49	76.77	9.76

- Boundary conditions.

### 1. Water transport and boundary conditions.

According to the experimental model, the upper boundary was in direct contact with the atmosphere and was set as the atmospheric boundary. The lower boundary was set as the variable head boundary due to the rise and fall of the water level.

### 2. Solute transport boundary.

According to the model, the upper boundary condition was the pollutant concentration boundary, and the lower boundary condition was the zero concentration gradient boundary. In the model setting, the primary considerations were adsorption and desorption, as well as nitrification and denitrification.

### 2.2.3. Model Parameters

The parameters of the numerical model mainly included soil hydraulic parameters and solute transport parameters. The initial values of the soil hydraulic parameters were determined according to the soil medium hydraulic parameter database of the Hydrus-1D software. The initial values of solute transport parameters were based on the measured results of this physical experiment, and the practical value was determined. The inverse solution module of the Hydrus-1D software was used for inversion to obtain the final parameters of the numerical model (Tables 3 and 4).

**Table 3.** Soil and water characteristic parameters.

Parameter	$\theta_r$ Residual Volume Water Content	$\theta_s$ Saturated Volume of Water Content	$\alpha/\text{cm}^{-1}$ Soil Moisture Characteristic Parameters	$n$ Soil Moisture Characteristic Index	$K_s/(\text{cm} \cdot \text{d}^{-1})$ Saturated Hydraulic Conductivity
Coarse sand	0.045	0.43	0.1450	2.68	712.8
Medium sand	0.051	0.42	0.1045	2.08	550.0
Fine sand	0.057	0.41	0.1240	2.28	350.2

**Table 4.** Solute transport parameters.

Medium	Solute	$\rho/\text{mg} \cdot \text{cm}^{-3}$ Bulk Density	$Disp/\text{cm}$ Longitudinal Diffusion	$D/\text{cm}^2 \cdot \text{d}^{-1}$ Solute Diffusion Coefficient	$K_d/\text{cm}^3 \cdot \text{mg}^{-1}$ Adsorption Coefficient
Coarse sand	Nitrite Nitrogen	1600	1.568	1.29085	0.000256
	Nitrate			1.69085	0.001584
	Ammonium Nitrogen			1.69085	0.008779
Medium sand	Nitrite Nitrogen	1800	1.233	1.29085	0.000103
	Nitrate			1.69085	0.000413
	Ammonium Nitrogen			1.69085	0.008895
Fine sand	Nitrite Nitrogen	1800	1.116	1.29085	0.000767
	Nitrate			1.69085	0.003094
	Ammonium Nitrogen			1.69085	0.008976

### 2.2.4. Calibration and Evaluation of the Model

The reliability was verified and analyzed by inputting the solute transport parameters and soil hydraulic parameters through the HYDRUS-1D software, and the simulation's accuracy was evaluated by the coefficient of determination  $R^2$ , and the root mean square error (RMSE). The closer the coefficient of determination  $R^2$  was to 1, the closer the root mean square error (RMSE) was to 0, which means that the model simulation results and the measured results of nitrogen had higher fitting accuracy. The calculation formula is:

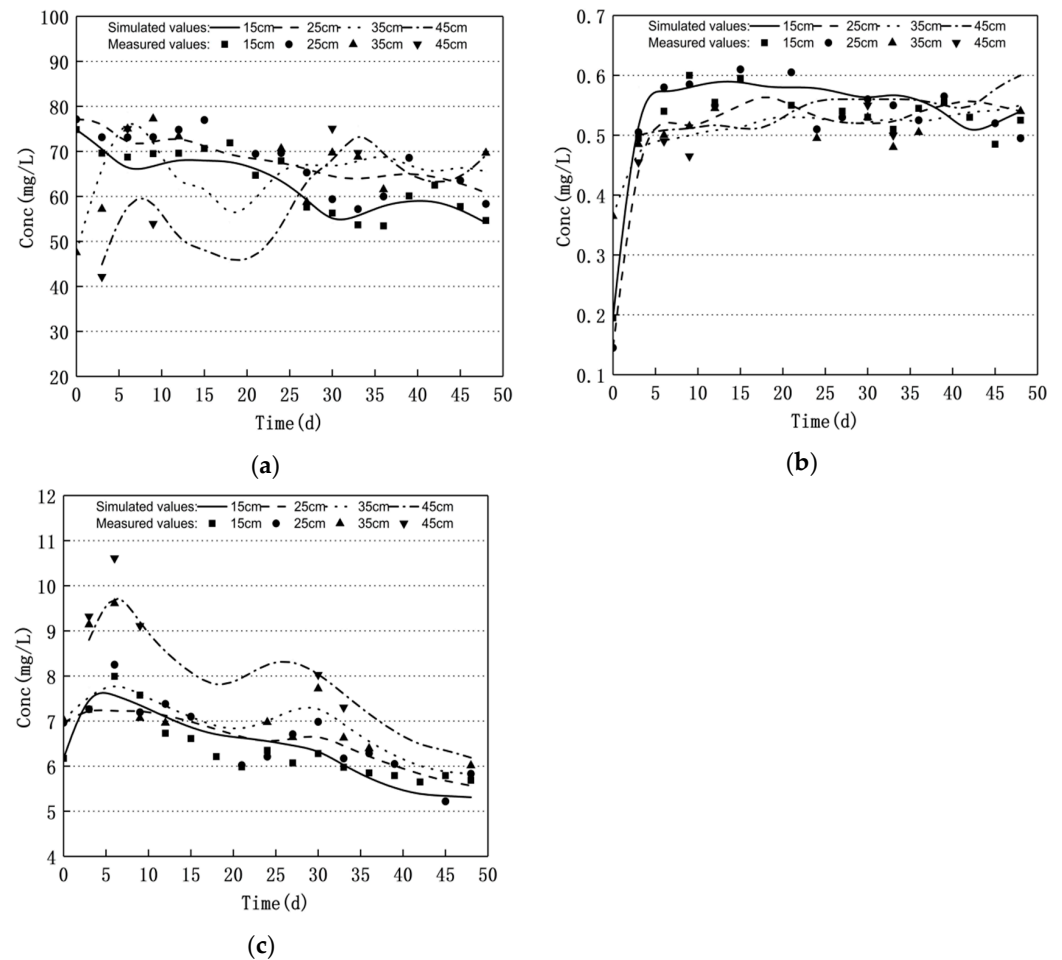
$$R^2 = \frac{\sum_{i=1}^n (\hat{y}_i - \bar{y})^2}{\sum_{i=1}^n (y_i - \bar{y})^2} \quad (5)$$

$$RMSE = \sqrt{\frac{1}{N} \sum_{i=1}^N (S_i - M_i)^2} \quad (6)$$

where  $S_i$  and  $M_i$  are simulated and measured values, respectively;  $N$  is the number of samples.

According to the simulation results of the numerical model of nitrogen migration and transformation in the water level fluctuation zone, the simulated values of ammonia nitrogen, nitrate nitrogen and nitrite nitrogen are shown in Figures 4–6, and the model fitting results are shown in Table 5. The correlation coefficients are mostly above 0.8.

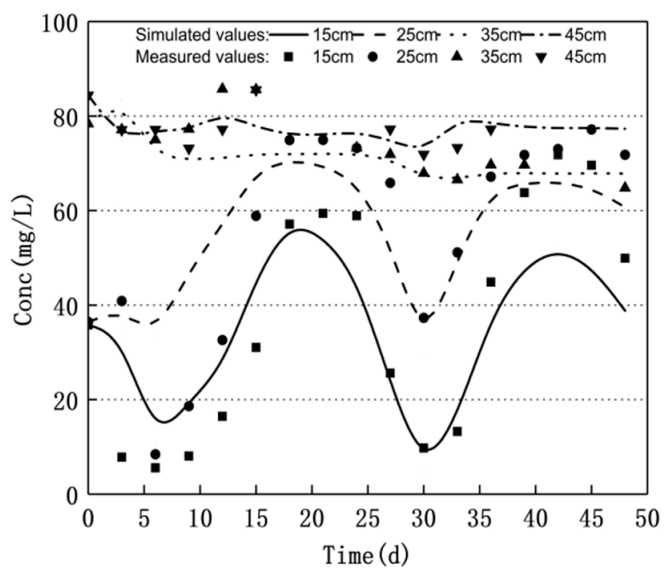
The root mean square error (*RMSE*) is small. Except for the significant simulation error of individual pollutants, the simulation results of the numerical model fit well with the measured results, indicating that the numerical model can better reflect the nitrogen transfer and transformation process under the fluctuation of the groundwater level.



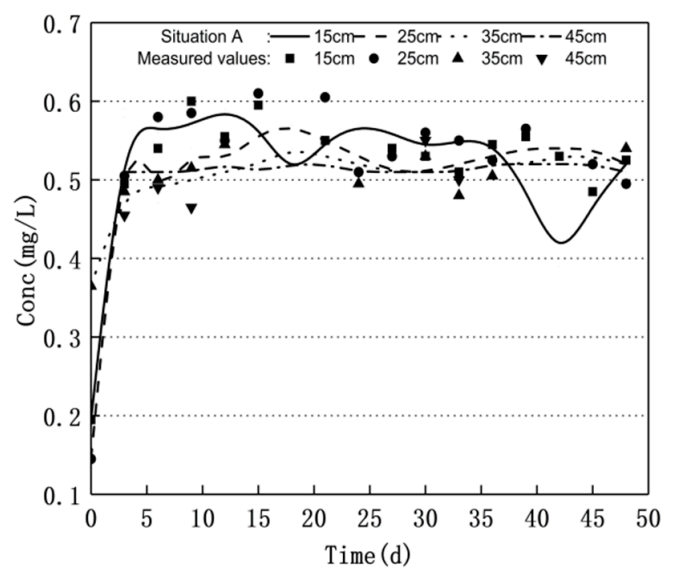
**Figure 4.** The process of nitrogen change in coarse sand ((a) nitrate nitrogen; (b) nitrite nitrogen; (c) ammonium nitrogen).

**Table 5.** Model fitting effect.

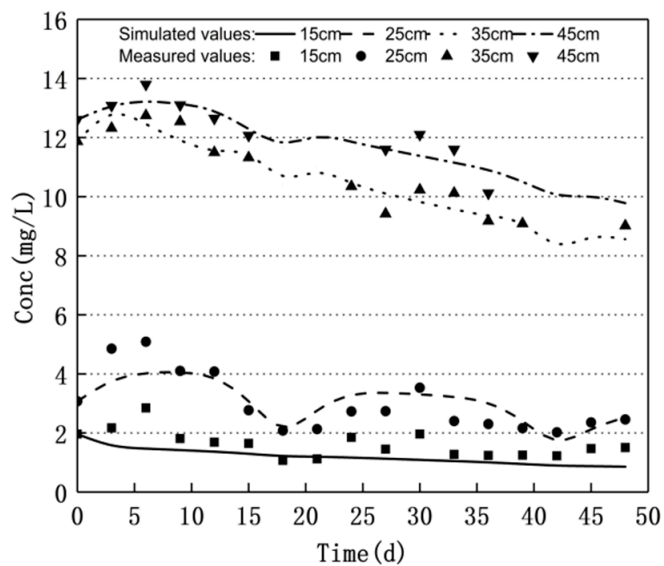
Soil Media	Pollutants	Decisive Factor $R^2$	$RMSE$
Coarse sand	Nitrite nitrogen	0.71284	0.0883
	Nitrate nitrogen	0.70572	5.4257
	Ammonia nitrogen	0.82099	0.4863
Medium sand	Nitrite nitrogen	0.74610	0.0486
	Nitrate nitrogen	0.80656	9.1317
	Ammonia nitrogen	0.98810	0.5115
Fine sand	Nitrite nitrogen	0.87036	0.0309
	Nitrate nitrogen	0.98398	2.7137
	Ammonia nitrogen	0.97552	0.3860



(a)



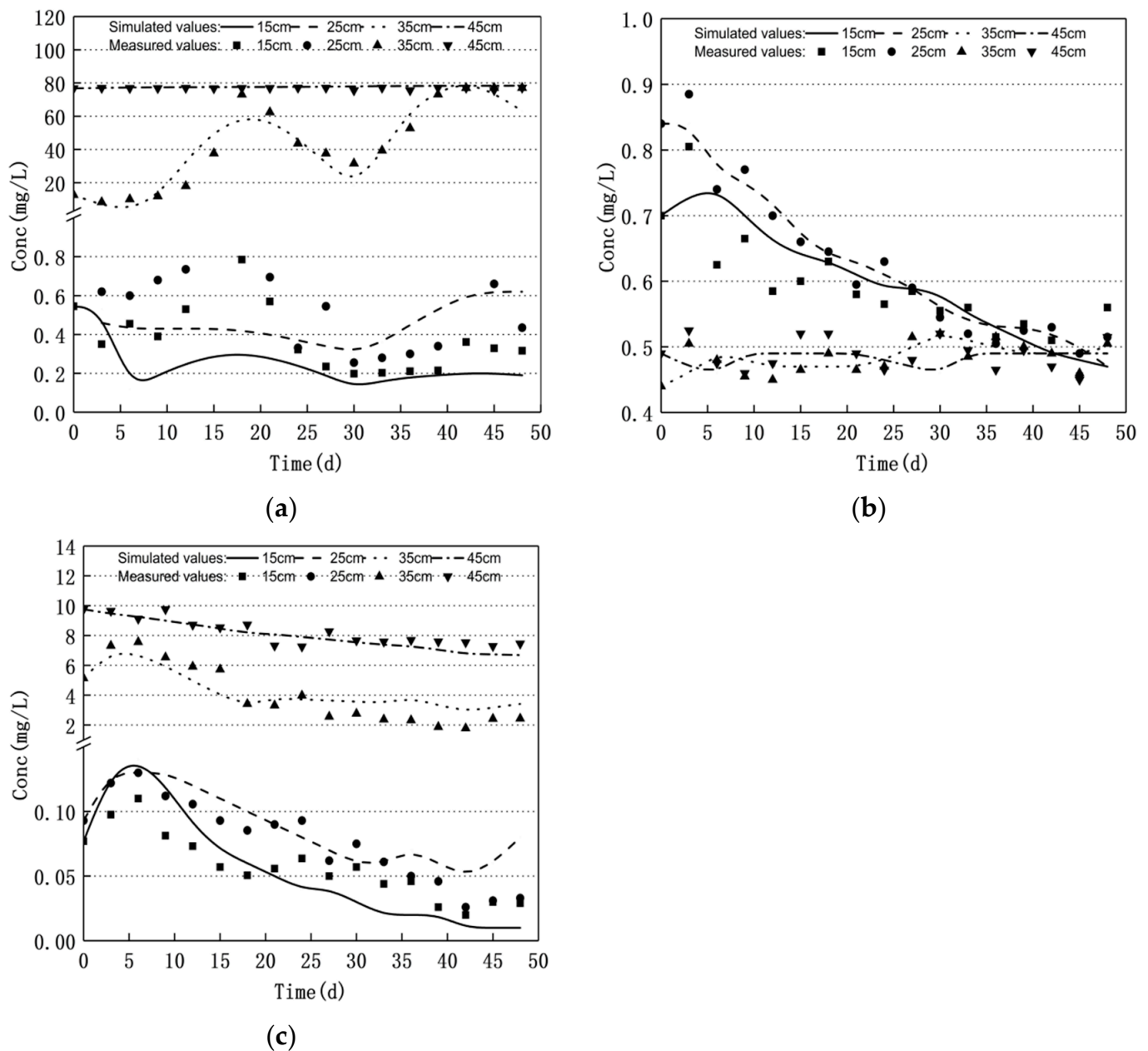
(b)



(c)

**Figure 5.** The process of nitrogen change in medium sand ((a) nitrate nitrogen; (b) nitrite nitrogen; (c) ammonium nitrogen).





**Figure 6.** The process of nitrogen change in fine sand ((a) nitrate nitrogen; (b) nitrite nitrogen; (c) ammonium nitrogen).

### 3. Results and Discussion

#### 3.1. Nitrogen Changes and Model Validation

The process curves of the measured and simulated results of ammonia nitrogen, nitrate nitrogen and nitrite nitrogen in the coarse sandy soil column under the condition of water level fluctuation are shown in Figure 4.

In the 15 cm and 25 cm sampling ports in the coarse sand column,  $\text{NO}_3^-$ -N showed a decreasing trend in the rising stage of the water level, with a decrease of 8.55% on average, the maximum decrease of 14.94% in the second cycle of the second rising stage of the water level, and the minimum decrease of 2.27% in the first rising stage of the second cycle of the water level. The concentration of  $\text{NO}_3^-$ -N showed an increasing trend in the declining water level stage, with an average increase of 9.10%, a maximum increase of 15.47% and a minimum increase of 4.62%, and the second fluctuation cycle was more obvious than the first fluctuation cycle of nitrate heel fluctuation. The average increase

of  $\text{NH}_4^+$ -N concentration was 9.16% at the stage of the water level rise, and the average decrease of  $\text{NH}_4^+$ -N concentration was 14.355% at the stage of water level fall, and the fluctuation change was more obvious in the first cycle. The concentration of  $\text{NO}_2^-$ -N was much smaller than that of the  $\text{NO}_3^-$ -N and  $\text{NH}_4^+$ -N concentrations, and  $\text{NO}_2^-$ -N showed fluctuating changes and eventually stabilized.

The process curves of the measured and simulated results of ammonia nitrogen, nitrate nitrogen and nitrite nitrogen in the medium sand soil column under the water level fluctuation conditions are shown in Figure 5.

In the medium sand column, the trend of  $\text{NO}_3^-$ -N was similar to that of the coarse sand soil column, with insignificant changes in concentrations at the 35 cm and 45 cm sampling ports, and obvious fluctuation trends at the 15 cm and 25 cm sampling ports. The concentration of  $\text{NO}_3^-$ -N at the 15 cm sampling port decreased by 780.30% on average, especially during the first water level rise in the first cycle. The concentration of  $\text{NO}_2^-$ -N was fluctuating at the beginning and stabilized later.

The process curves of the measured and simulated results of ammonia nitrogen, nitrate nitrogen and nitrite nitrogen in the fine sandy soil column under the condition of water level fluctuation are shown in Figure 6.

In the fine sand soil column, the trend of the  $\text{NO}_3^-$ -N concentration changes at the 15 cm, 25 cm and 35 cm sampling ports were basically the same, and the decreases at 15 cm, 25 cm and 35 cm were 25.93%, 68.05% and 19.19, respectively, during the water level rise stage, and the increases were at 15 cm, 25 cm and 35 cm during the water level fall stage. The trends of  $\text{NH}_4^+$ -N concentrations at the 15 cm, 25 cm and 35 cm sampling ports were basically the same, and the concentrations at the 45 cm sampling port did not change much, and the increases at the water level rising stage were 32.44%, 23.39% and 27.03% for 15 cm, 25 cm and 35 cm, respectively, and the decreasing water level for the  $\text{NO}_2^-$ -N concentration was low and stabilized after fluctuating changes.

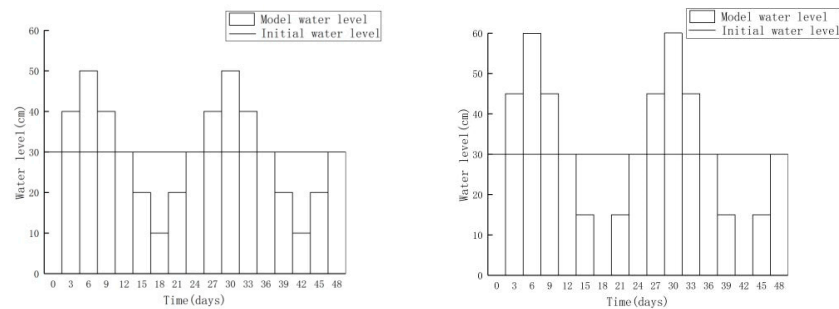
Figures 4–6 show the actual measurement process curve of ammonia nitrogen, nitrate nitrogen and nitrite nitrogen under the conditions of water level fluctuation. When the water level rises, the dissolved oxygen content decreases, and the  $\text{NH}_4^+$ -N concentration should fall. Still, denitrifying bacteria become active and dominant under hypoxic conditions, promoting the increase in the  $\text{NH}_4^+$ -N concentration and the significant decrease in the  $\text{NO}_3^-$ -N concentration. When the water level drops, the dissolved oxygen content increases and nitrification plays a central role. The concentration of  $\text{NO}_3^-$ -N increases significantly. However, due to the strong adsorption of the soil, the concentration of  $\text{NH}_4^+$ -N in the free water of the soil solution decreases [43,44]. Nitrite nitrogen, in the three media, fluctuated and eventually stabilized, but its concentration was much smaller than the concentrations of  $\text{NO}_3^-$ -N and  $\text{NH}_4^+$ -N. Before the water level rises, when the dissolved oxygen is sufficient, the reproduction rate of nitrifying bacteria is slower than that of nitrosating bacteria; at this time, the nitrosation reaction dominates, causing the accumulation of  $\text{NO}_2^-$ -N, resulting in an increased concentration of  $\text{NO}_2^-$ -N in the soil solution during the rising water level stage. During the falling phase of the water level, nitrification dominates, a lot of  $\text{H}^+$  is produced in the solution, and the soil solution becomes weakly acidic, which strengthens the conversion of nitrite to nitrate.  $\text{NO}_2^-$ -N no longer accumulates, and the concentration gradually decreases [29,45,46]. In the initial stage, the increase in  $\text{NO}_2^-$ -N satisfies coarse sand, then medium sand, then fine sand. The more significant the particle size is, the more particle surfaces there are that microbial flocs can come into contact with, and more microbes can participate in the nitrification reaction. With a stronger microbial nitrification ability, it is understood that the nitrification ability of microbial flocs will increase with the increase in particle size [47].

### 3.2. Scenario Simulation

#### 3.2.1. Increasing Water Level Fluctuation

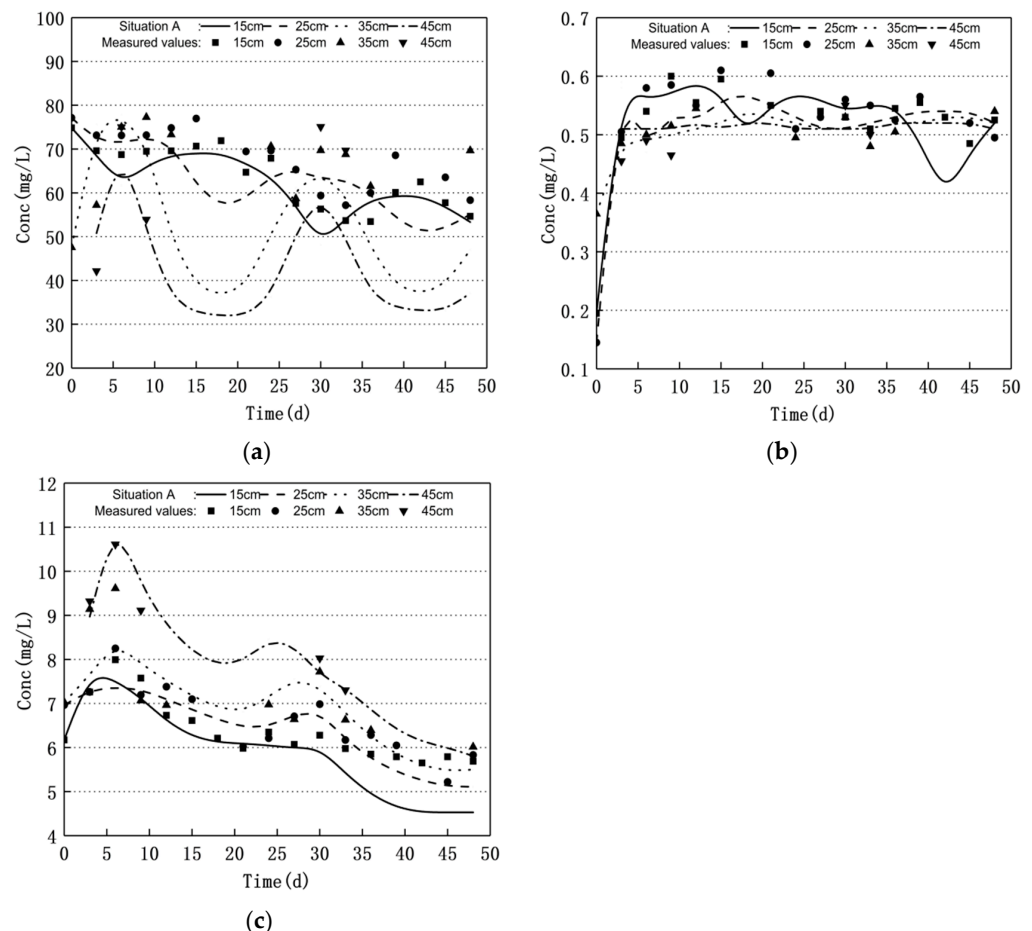
Scenario 1 is set to: keep the initial pollutant concentration unchanged, and increase the water level fluctuation range. That is, the initial water level is set to 30 cm, and the

height is raised by 15 cm at a time, and then rises twice until the water level reaches 60 cm; then it is dropped by 15 cm each time and drops four times until the water level drops to 0 cm; finally, the water level is raised again twice, by 15 cm each time, until it reaches 30 cm, which is the initial water level. The comparison chart of water level changes is shown in Figure 7, recorded as situation A.

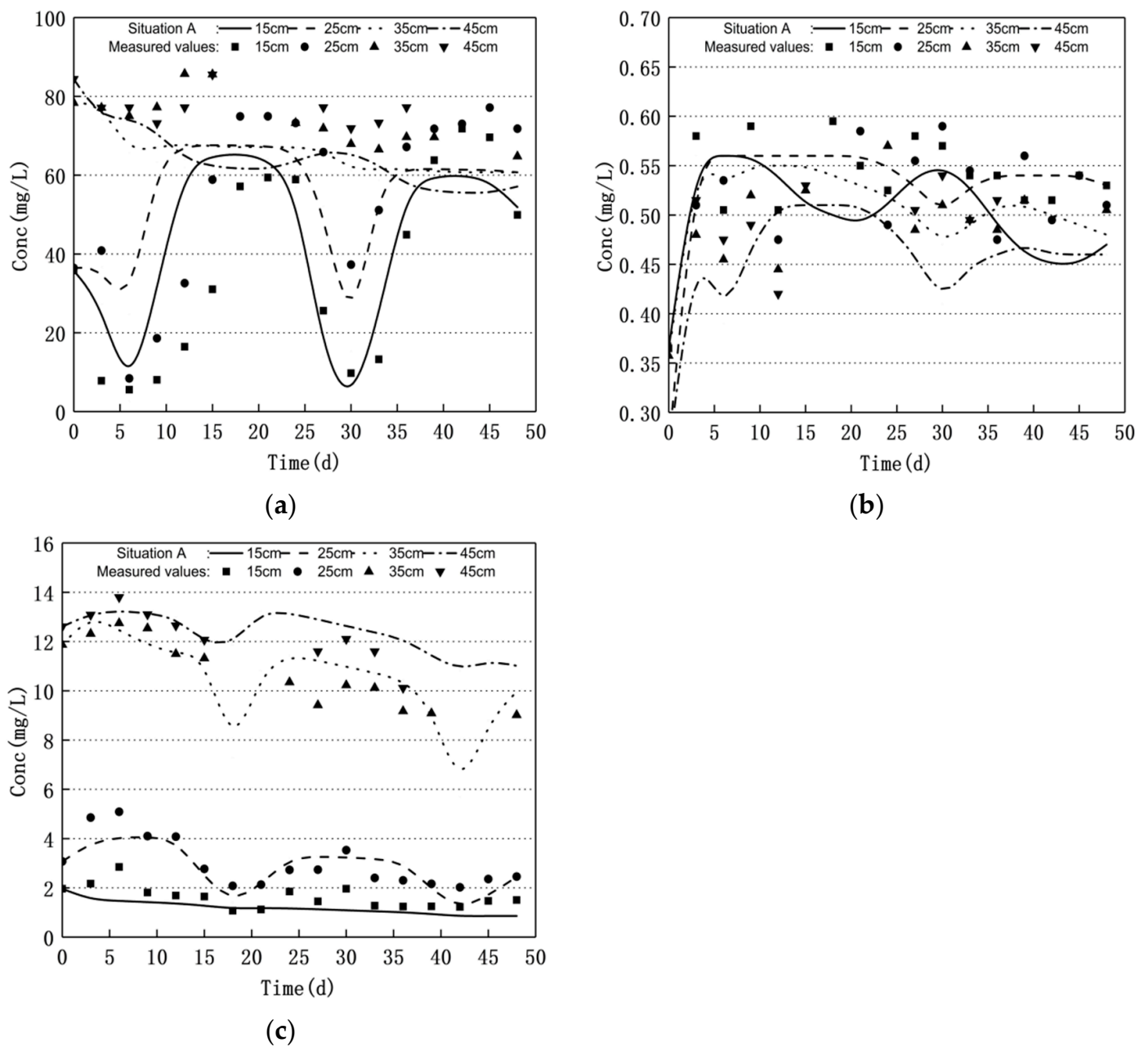


**Figure 7.** Comparison of water level changes.

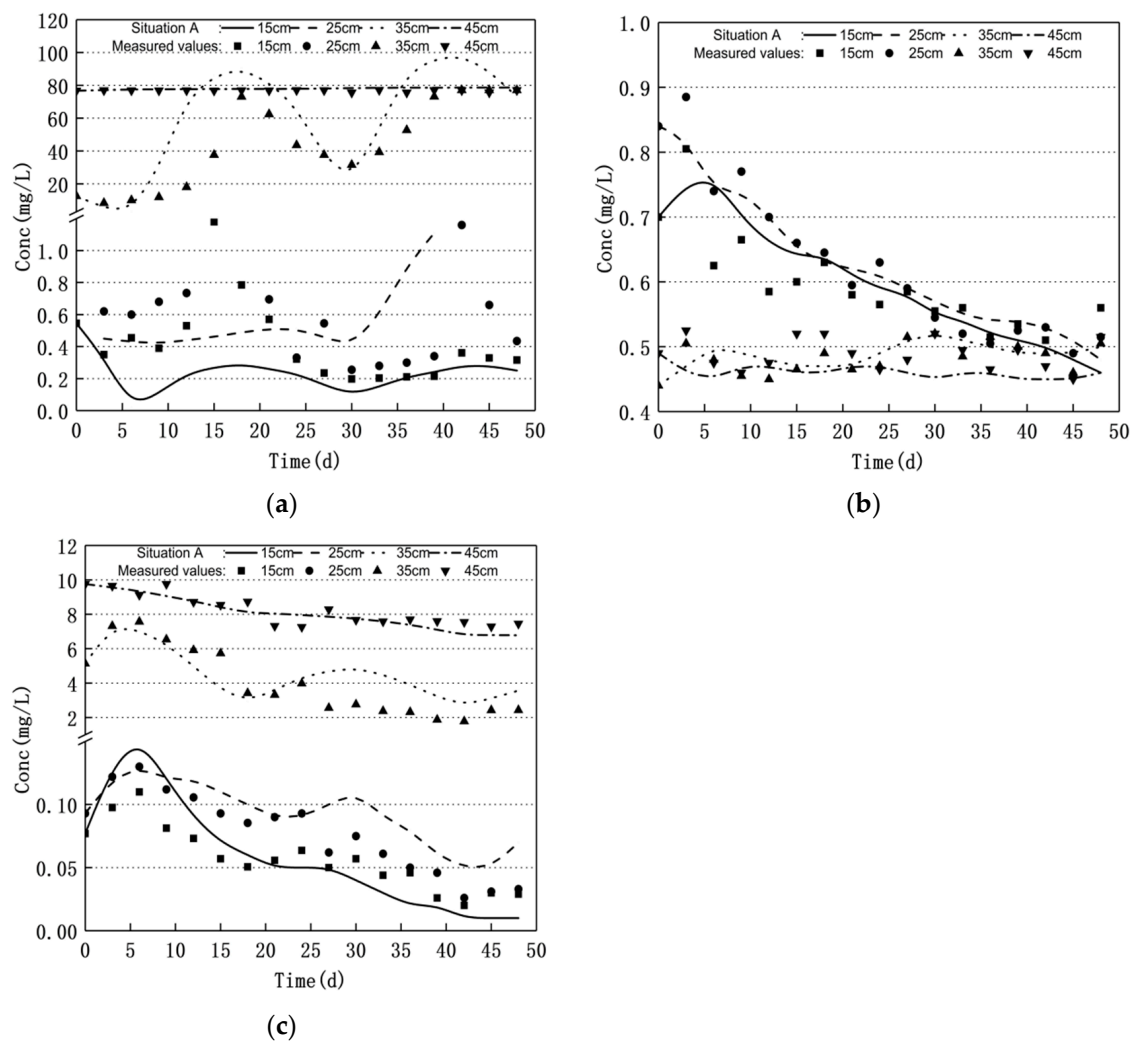
Synchronously with the previous experiment scenario of groundwater level fluctuation, nitrogen migration and transformation (henceforth referred to as the fluctuation experiment), the simulation period is set to two by increasing the amplitude of the water level fluctuation, and the range of solute concentration in the water body increases. The model is built and run according to the scenario, and the simulation result is shown in Figures 8–10.



**Figure 8.** The measured results of coarse sand and the process of dynamic change of nitrogen in situation A ((a) nitrate nitrogen; (b) nitrite nitrogen; (c) ammonium nitrogen).



**Figure 9.** The measured results of medium sand and the dynamic process of nitrogen in situation A ((a) nitrate nitrogen; (b) nitrite nitrogen; (c) ammonium nitrogen).



**Figure 10.** The measured results of fine sand and the dynamic process of nitrogen in situation A ((a) nitrate nitrogen; (b) nitrite nitrogen; (c) ammonium nitrogen).

The comparison of the standard deviation of nitrogen concentration obtained by the three media simulations is shown in Table 6.

**Table 6.** Scenario A model standard deviation comparison.

	Observation Hole	Coarse Sand		Medium Sand		Fine Sand	
		Fluctuation Experiment	Situation A	Fluctuation Experiment	Situation A	Fluctuation Experiment	Situation A
Nitrite nitrogen	15 cm	0.0249	0.0488	0.0348	0.0384	0.0838	0.0896
	25 cm	0.0197	0.0262	0.0141	0.0168	0.1037	0.0924
	35 cm	0.0159	0.0135	0.0261	0.0263	0.0166	0.0159
	45 cm	0.0276	0.0047	0.0255	0.0328	0.0107	0.0072
Nitrate	15 cm	5.4152	6.2294	16.0273	22.4563	0.0921	0.0807
	25 cm	4.2269	6.9961	12.8282	15.5376	0.0932	0.3326
	35 cm	5.2402	14.0076	3.8663	4.4202	24.2231	31.5546
	45 cm	9.5118	11.4918	1.9142	6.0021	0.417	0.4754
Ammonium Nitrogen	15 cm	0.7797	1.0168	0.2074	0.2113	0.0422	0.0441
	25 cm	0.5452	0.7755	0.7561	0.9278	0.027	0.0248
	35 cm	0.6154	0.875	1.4036	1.8423	1.2271	1.3702
	45 cm	1.0365	1.4005	1.1279	0.8256	0.8747	0.8622



Table 6 shows that the increase in the fluctuation range of water level can effectively enlarge solute fluctuation.

Water level fluctuations have different effects on the fluctuation range of nitrogen concentration in the three media. When the fluctuation range of water level increases by 5 cm, the fluctuation range of nitrogen in the coarse sand medium increases by 37.52% on average, compared to the fluctuation experiment scenario; the nitrogen concentration in the medium sand medium is increased by 37.52%. The fluctuation range increased by 31.40% on average; the fluctuation range of the nitrogen concentration in the fine sand medium increased by 21.14% on average.

Table 6 shows that in the coarse sand, the fluctuation of the nitrogen concentration changes most significantly with the increase in the fluctuation range of the water level, followed by the medium sand, and the fine sand has the slightest change.

The impact of water level fluctuations on the three solutes is also different. The fluctuation range of the water level is expanded by 5 cm, and the fluctuation range of nitrite nitrogen, nitrate nitrogen and ammonium nitrogen in the coarse sand medium increases by 7.90%, 67.17%, and 37.49%, respectively. In the medium sand medium, the fluctuation range increased by 14.66%, 72.28%, and 7.26%, respectively. The fluctuation range of nitrite nitrogen in the fine sand medium decreased by 10.34%, while the fluctuation range of nitrate nitrogen and ammonium nitrogen increased by 72.17% and 1.62%, respectively.

Table 6 shows that among the three media, the fluctuation of the water level has the most significant effect on the fluctuation of nitrate nitrogen, and the influence on the fluctuation of nitrite nitrogen and ammonium nitrogen is relatively small.

### 3.2.2. Scenario of Reduced Water Level Fluctuation

Scenario 2 is set to: keep the initial pollutant concentration unchanged, and reduce the fluctuation of the water level. That is, the initial water level is set to 30 cm, and the height is raised by 5 cm each time, and then rises two times until the water level reaches 40 cm; it then starts to drop by 5 cm each time, and drops four times until the water level drops to 20 cm; finally, the water level rises again two times, by 5 cm each time, until it reaches 30 cm, which is the initial water level. The comparison chart of the water level changes is shown in Figure 11. It is recorded as situation B.

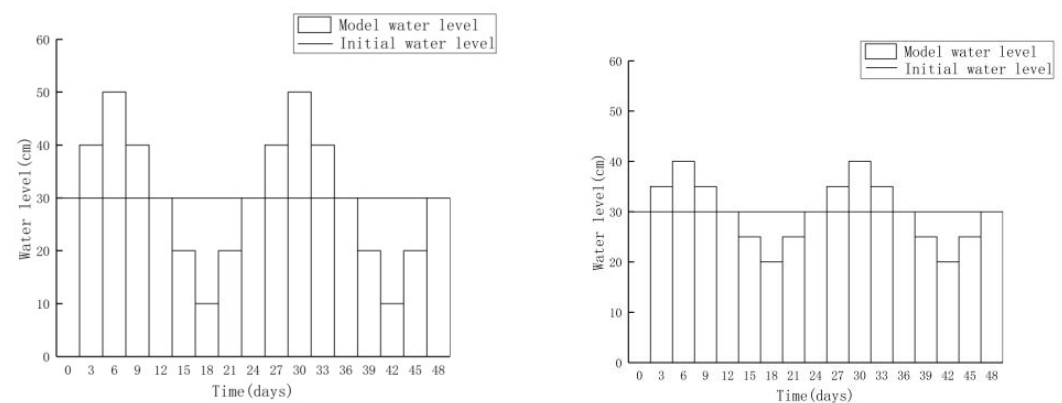
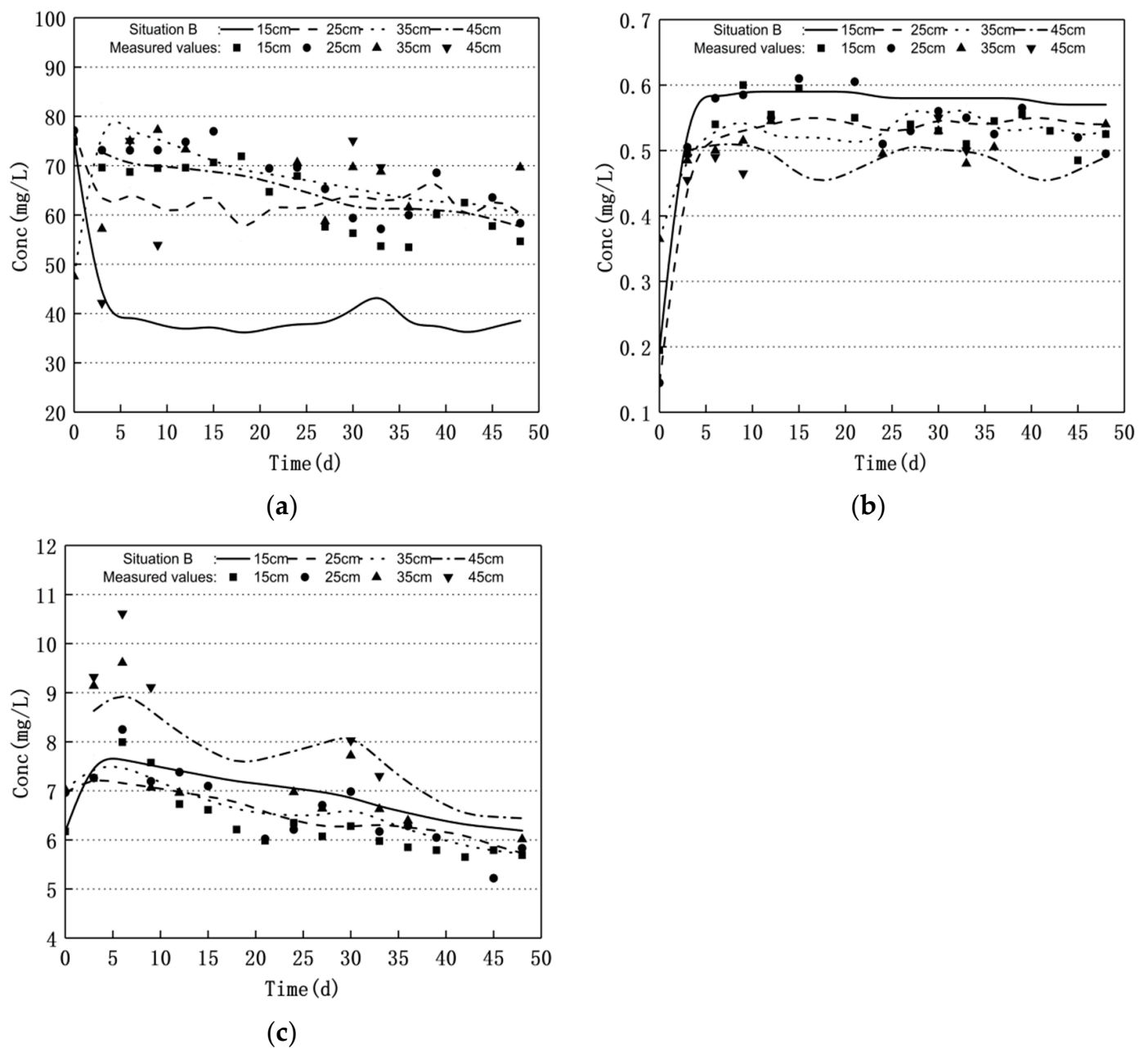
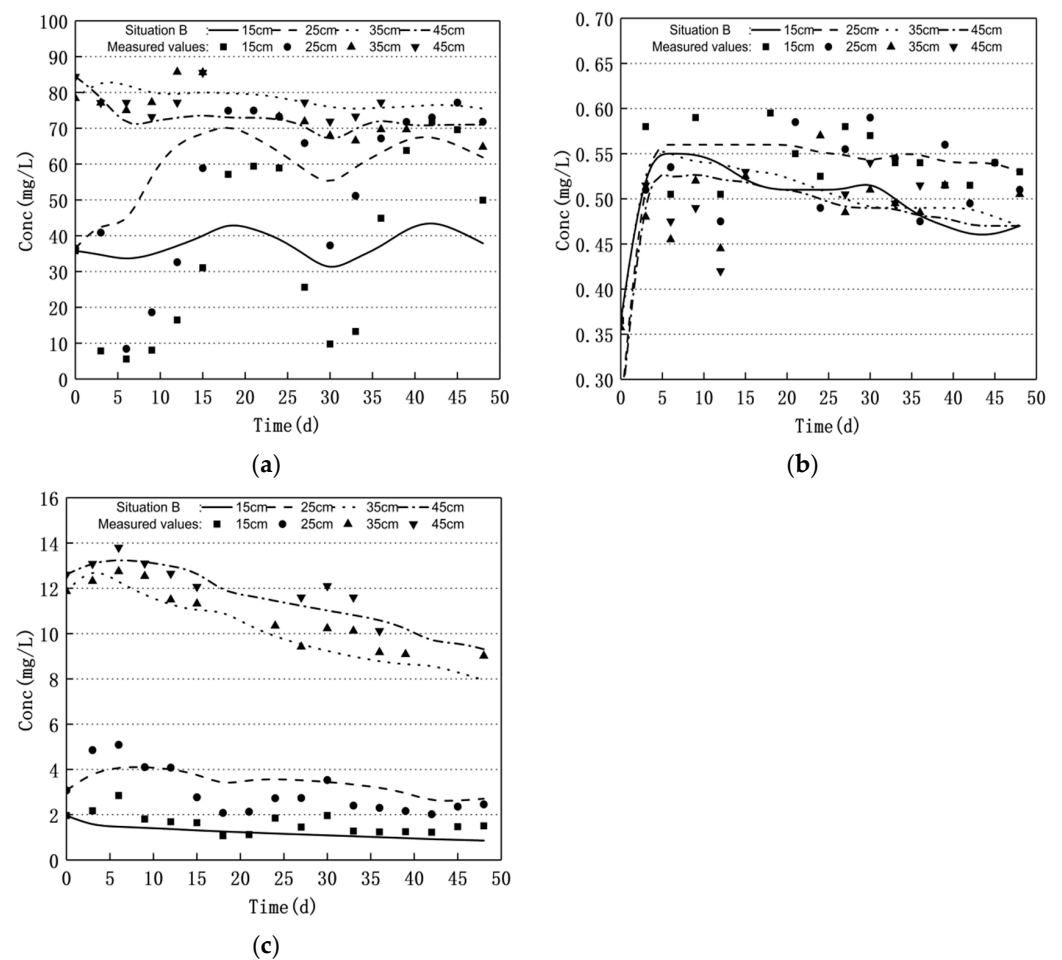


Figure 11. Comparison of Water Level Changes.

Similarly, the simulation period is set to 2. By increasing the fluctuation range of the water level, the variation range of the solute concentration in the water body is increased. The model is built and run according to the scenario, and the simulation result is shown in Figures 12–14.



**Figure 12.** The measured results of coarse sand and the process of dynamic change of nitrogen in situation B ((a) nitrate nitrogen; (b) nitrite nitrogen; (c) ammonium nitrogen).

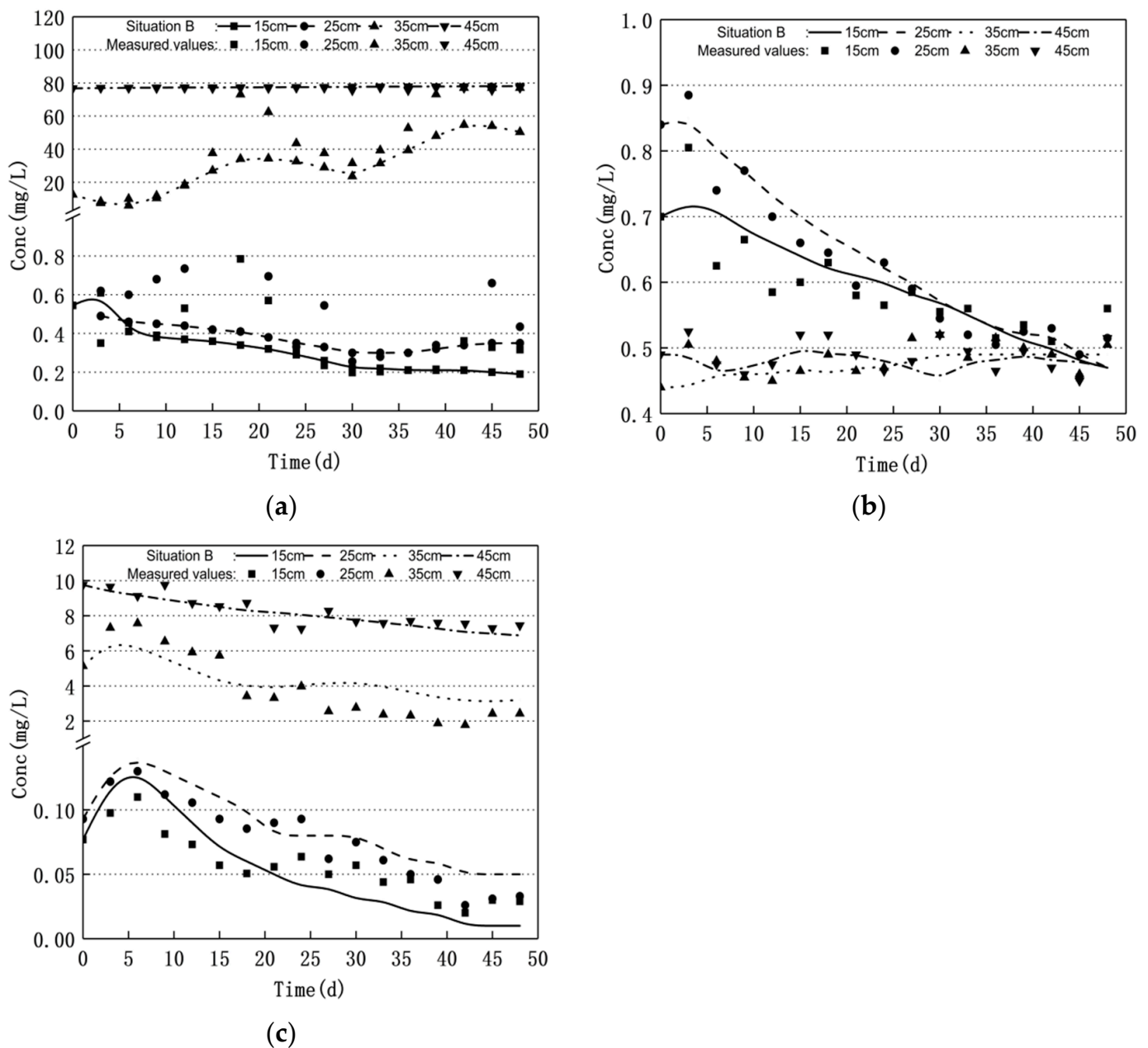


**Figure 13.** The measured results of medium sand and the dynamic process of nitrogen in situation B ((a) nitrate nitrogen; (b) nitrite nitrogen; (c) ammonium nitrogen).

The comparison of the standard deviation of the nitrogen concentration obtained by the three media simulations is shown in Table 7.

**Table 7.** Scenario B model standard deviation comparison.

	Observation Hole	Coarse Sand		Medium Sand		Fine Sand	
		Fluctuation Experiment	Situation B	Fluctuation Experiment	Situation B	Fluctuation Experiment	Situation B
Nitrite Nitrogen	15 cm	0.0249	0.0063	0.0348	0.0316	0.0838	0.0758
	25 cm	0.0197	0.0143	0.0141	0.0099	0.1037	0.1139
	35 cm	0.0159	0.0183	0.0261	0.0266	0.0166	0.0148
	45 cm	0.0276	0.0206	0.0255	0.0219	0.0107	0.0112
Nitrate	15 cm	5.4152	2.1756	16.0273	4.0429	0.0921	0.1069
	25 cm	4.2269	3.337	12.8282	8.1699	0.0932	0.0601
	35 cm	5.2402	6.0552	3.8663	2.4465	24.2231	15.1679
	45 cm	9.5118	4.4877	1.9142	2.658	0.417	0.3406
Ammonium Nitrogen	15 cm	0.7797	0.4898	0.2074	0.2042	0.0422	0.0391
	25 cm	0.5452	0.4511	0.7561	0.4871	0.027	0.029
	35 cm	0.6154	0.5448	1.4036	1.4785	1.2271	1.0044
	45 cm	1.0365	0.7817	1.1279	1.3127	0.8747	0.7587



**Figure 14.** The measured results of fine sand and the dynamic process of nitrogen in situation B ((a) nitrate nitrogen; (b) nitrite nitrogen; (c) ammonium nitrogen).

Table 7 shows that the decrease in water level fluctuation can effectively reduce solute fluctuation.

Water level fluctuations have different effects on the fluctuation range of nitrogen concentration in the three media. When the fluctuation range of the water level decreases by 5 cm, the fluctuation range of nitrogen in the coarse sand medium is reduced by 36.74% on average, compared to the fluctuation experiment scenario. In the medium sand medium, the fluctuation range of the concentration of nitrogen decreased by 14.70% on average, while the fluctuation range of the nitrogen concentration in the fine sand medium decreased by 9.39% on average.

Table 7 shows that in the coarse sand, the fluctuation of the nitrogen concentration changes most significantly with the decrease in the fluctuation range of the water level, followed by the medium sand, and the fine sand demonstrates the most minor change.

The impact of water level fluctuations on the three solutes is also different. When the fluctuation range of the water level is reduced by 5 cm, the fluctuation ranges of nitrite nitrogen, nitrate nitrogen and ammonium nitrogen in the coarse sand medium are reduced by 28.07%, 29.55%, and 22.62%, respectively. In the medium sand medium, the average fluctuation range is reduced by 13.00%, 27.23%, and 3.85%, respectively. In the fine sand model, the fluctuation range is reduced by an average of 1.51%, 18.79%, and 7.86%, respectively.

Table 7 shows that among the three media, water level fluctuations have the most significant impact on the fluctuations of nitrate nitrogen, have less impact on the fluctuations of nitrite nitrogen and ammonium nitrogen, and the difference is the most obvious in the fine sand medium, followed by medium sand. This difference is not great in the coarse sand.

#### 4. Conclusions

In this paper, through the experiment examining nitrogen migration and transformation in the groundwater fluctuating zone, we analyzed the nitrogen migration and transformation process. A numerical model of nitrogen migration and transformation in the groundwater level fluctuating zone was established with the help of the HYDRUS-1D model. The paper obtained the following main conclusions:

- Groundwater level fluctuations can significantly affect the nitrogen transport and transformation patterns in soil–groundwater. The nitrate nitrogen concentration increased and the ammonium nitrogen mass concentration decreased when the water level decreased. Moreover, the nitrate nitrogen mass concentration decreased and the ammonium nitrogen mass concentration increased when the water level increased; the nitrite nitrogen did not change significantly.
- In this study, indoor soil column experiments were combined with the Hydrus-1D software simulation prediction to simulate the theoretical values of the tri-nitrogen transformation process in indoor soil columns. Although simple, the software does not take into account the existence of non-homogeneous changes in the soil column, is not accurate enough, there are certain limitations, and the influence of biological processes on the transformation of pollutants has not been taken into account. Furthermore, there is still a certain gap with the actual results.
- As an important factor of the hydrological mechanism, the water level plays a vital role in the groundwater system, and the effect of the level and intensity of fluctuation of the groundwater level on the migration and transformation of nitrogen cannot be ignored. The change in water level will affect the water content in the soil, the state of water movement, the physical and chemical properties of soil and the state of the microorganisms, which in turn, will affect the migration and transformation of nitrogen in the soil.

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